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Determining Plutonium Mass in Spent Fuel with Nondestructive Assay Techniques - Preliminary Modeling Results Emphasizing Integration among Techniques

by

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Determining Plutonium Mass in Spent Fuel with Nondestructive Assay Techniques - Preliminary Modeling Results Emphasizing Integration among Techniques

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Abstract – *There are a variety of motivations for quantifying Pu in spent (used) fuel assemblies by means of nondestructive assay (NDA) including the following: strengthen the capabilities of the International Atomic Energy Agencies to safeguards nuclear facilities, quantifying shipper/receiver difference, determining the input accountability value at reprocessing facilities and providing quantitative input to burnup credit determination for repositories. For the purpose of determining the Pu mass in spent fuel assemblies, twelve NDA techniques were identified that provide information about the composition of an assembly. A key point motivating the present research path is the realization that none of these techniques, in isolation, is capable of both (1) quantifying the elemental Pu mass of an assembly and (2) detecting the diversion of a significant number of pins. As such, the focus of this work is determining how to best integrate 2 or 3 techniques into a system that can quantify elemental Pu and to assess how well this system can detect material diversion. Furthermore, it is important economically to down-select among the various techniques before advancing to the experimental phase. In order to achieve this dual goal of integration and down-selection, a Monte Carlo library of PWR assemblies was created and is described in another paper at Global 2009 (Fensin et al.). The research presented here emphasizes integration among techniques. An overview of a five year research plan starting in 2009 is given. Preliminary modeling results for the Monte Carlo assembly library are presented for 3 NDA techniques: Delayed Neutrons, Differential Die-Away, and Nuclear Resonance Fluorescence. As part of the focus on integration, the concept of "Pu isotopic correlation" is discussed and the role of cooling time determination.*

I. INTRODUCTION

Although the majority of plutonium (Pu) in the world is stored in commercial spent fuel assemblies, a measurement system for directly quantifying the Pu mass contained in these assemblies does not exist. The nondestructive assay systems in use today (Safeguards Mox Python Detector,[1] Fork Detector[2] and Cerenkov Viewing Device [3]) primarily measure indirect signatures from spent fuel such as gamma emission from fission fragments, or photons induced by radiation from fission fragment, or total neutron emission pre-dominantly emitted from curium. Calculation codes, known as burnup codes, can be used to infer plutonium mass from these measured signatures. In order to use burnup codes to predict the Pu mass in a particular assembly, input from the operator is required. From an international safeguards perspective, this input is undesirable given the regulatory requirement of independent verification.

Below, nine reasons for improving on the status quo are listed. These reasons are the motivation for designing a nondestructive assay (NDA) system that can quantify the Pu mass in spent fuel assemblies: (1) Provide regulators with the capability to independently verify the mass of plutonium at any site that has spent fuel. (2) Enable regulators and facilities to accurately quantify the Pu mass leaving one facility and arriving at another facility ("shipper/receiver difference"). (3) Provide confidence to the public that the shipment of spent fuel around the world is being undertaken in a rigorous manner; assure that material is not diverted during shipment. (4) Provide regulators with a tool for recovering continuity of knowledge at any site storing spent fuel. (5) Provide reactor operators with a tool enabling optimal reloading of reactor cores. (6) Provide regulators of once-through fuel cycle

repositories the capability to optimally pack fuel both for transport, in a pool and into the repository ("burnup credit"). (7) Enable determination of the input accountability mass of an electro-chemical (pyro-chemical) processing facility. (8) Provide facility operators with a means for quantifying the Pu mass in spent fuel that is no longer considered "self-protecting." This is particularly relevant given that some regulatory agencies are considering changes to the level at which radioactive material is considered to be self-protecting. And (9) promote cost savings by facilitating assembly selection for reprocessing. Facility operators blend assemblies to obtain optimal chemical compositions in reprocessing solutions. The blending is presently based on reactor history and burnup codes. The inaccuracy of the status quo decreases plant operational efficiency.

II. THE NEED FOR INTEGRATION

With the goal of quantifying the Pu mass in spent fuel assemblies, researchers identified twelve NDA techniques that quantify various signatures from commercial spent fuel.[4] The approach for researching the capabilities of these techniques was shaped by two key factors: (1) None of the NDA techniques is capable of determining elemental Pu mass as a standalone technique. And (2) several different NDA systems will likely be needed to satisfy the unique situations of the 9 motivations listed above; to expand on this point, factors such as cost, accuracy, and portability will impact what system of techniques are best for a given motivation.

The 12 NDA techniques being researched are the following: Delayed Neutrons[5], Differential Die-Away[6], Lead Slowing Down Spectrometer,[7,8,9,10] Neutron Multiplicity,[11] Passive Neutron Albedo Reactivity,[12,13,14] Total Neutron (Gross Neutron),[15] X-Ray Fluorescence,[16,17,18] ^{252}Cf Interrogation with Prompt Neutron Detection,[19] Delayed Gamma,[15] Nuclear Resonance Fluorescence,[20] Passive Prompt Gamma,[15] Self-integration Neutron Resonance Densitometry.[21,22]

III. TECHNICAL APPROACH

The research plan to determine Pu mass in spent fuel that started in early 2009 is nominally a five year effort and is part of the Next Generation Safeguard Initiative.[23] The initial two years are focused primarily on Monte Carlo modeling while the later three years involve the fabrication of hardware and measuring spent fuel assemblies. The Monte Carlo effort has two main goals: (1) Quantify the expected capability of each technique as an independent instrument; the performance of each NDA technique will be documented in 12 independent reports. (2) Determine how to integrate a few techniques together in order to determine elemental Pu mass. The result of the integration will be documented in one or more reports. In order to cost-effectively and robustly achieve these two goals, a library of assemblies was created.

In order to keep the assembly library manageable in size, one reactor type (pressurized water) was selected. The largest standard assembly size (17 by 17) was selected given that larger assemblies are generally more challenging than small ones. The differences among the assemblies emphasized isotopic, spatial and diversion variability as described in the next three paragraphs.

The isotopic variability among 64 assemblies was obtained by using the Monte Carlo N-Particle eXtended (MCNPX) transport code that recently had the CINDER burnup capability added.[24] Each assembly has a unique combination of burnup, enrichment, and cooling time. The burnup cases were 15, 30, 45 and 60 GWd/tU; the initial enrichments were 2, 3, 4, 5%; and the cooling times were 1, 5, 20, 80 years.

Since integration among techniques is an upfront requirement, it was necessary to assure that the assembly library contained technique-specific physical attributes. For example, the 100 keV photons measured with X-Ray Fluorescence come primarily from the outer ~200 μm of each pin. Over this same dimension, the plutonium content can vary by over a factor of two. As such, it is necessary to burn the fuel with radial resolution in every pin. This was done by dividing the fuel into 4 radial cells emphasizing the exterior portion of the pin. Another example of including technique-specific properties in the spent fuel library involves the hydrogen sensitivity of the Lead Slowing Down Spectrometer. The level of hydrogen in the fuel cladding produces a noticeable effect; and so the variation in the hydrogen content in the cladding as a function of burnup was included.

Over 40 "diversion assemblies" were created from a few of the 64 "non-diversion" assemblies. The majority of the diversion assemblies involve replacing pins from the center, mid and outer regions with natural uranium pins. The details of the diversion assemblies are presented in a paper by Fensin et al. at this conference.[25]

The performance of each instrument will be quantified for the full 100+ assemblies in the library as if the measurements took place in three different media: air, water and borated water with the exception of the Lead Slowing Down Spectrometer which cannot operate in water. In total, over 3,000 models will be run with the assemblies of the library.

The ideal situation for the experimental phase of determining Pu mass in spent fuel is to compare the Pu mass determined with NDA for an individual assembly to the Pu mass determined from an input accountability tank (IAT) for that same individual assembly. Given that most IATs are not made to handle one assembly and given that there are few facilities with IATs able to be spent for research, it is likely that this ideal situation will not be

satisfied. It is hoped that a less than ideal situation will provide a proof-of-principle to one or two of the final NDA system designs.

Some of measurement options that may be possible include the following: (1) Compare the average Pu mass of ~10 assemblies in an IAT to the NDA predicted mass of the ~10 assemblies. For this path, if a continuous dissolver is spent, attention will need to be taken to assure that mass from other assemblies is not added to the IAT nor that mass from the 10 assemblies measured with NDA is not lost from the IAT. A batch dissolver would be preferable. (2) Determine the Pu mass in an IAT with "x" assemblies dissolved in it and then repeat for "x+1" assemblies. For this approach, the issues described above with continuous dissolvers are still valid. (3) Combining DA of several pellets in an assembly with burnup codes in order to render a "best possible estimate" of the Pu mass of an assembly in the absence of an IAT. (4) Comparing NDA measurements of assemblies to well bench marked burnup codes. Collaborators interested in participating in the experimental, or modeling work, are encourage to contact us.

IV. INTEGRATION OVERVIEW

Depending on the customers of an NDA system capable of measuring Pu mass in spent fuel assemblies, the design of the system will vary. For example, the needs of the International Atomic Energy Agency (IAEA) inspectors at a reactor site vary from those of IAEA inspectors at repository or reprocessing facilities. Furthermore, the needs of domestic regulators or facility operators will vary from those of IAEA inspectors. Although this is true, there are a number of aspects and potential analysis approaches that will likely be similar to several systems.

Since the diversion of pins is a central concern for IAEA inspectors, any system needing to detect such diversion will have one of 6 neutron-based techniques as part of the system – in particular one of the first 6 instruments listed at the end of section II above. This is because each of these six techniques is expected to be sensitive to pin removal anywhere in the assembly. This is primarily due to the greater penetrating ability of neutrons as well as the propagation of neutrons through multiplication. In contrast, all of the photon techniques are expected to be blind to pin removal from the interior of the assembly.

The next three sections describe the capability of three different NDA techniques. These techniques were selected since they are in the process of being investigated in the context of the spent fuel library. Detailed reports on each will be completed within approximately a year.

V. DELAYED NEUTRON DETECTION

Delayed neutron techniques involve active interrogation. The fissile content is measured with an emphasis on the ^{235}U content. When assaying spent fuel ~2.1 times more delayed neutrons are emitted from ^{235}U than from Pu on a per fission basis. The isotopic ratio between ^{239}Pu and ^{241}Pu spent for this determination is representative of a ~40 GWd/tU, 4% enriched, 5 years cooled assembly.

The basic concept of using delayed neutrons in the context of spent fuel is the following: (1) An external neutron source produces neutrons near the assembly. These neutrons induce fissions, (2) The external neutron source is removed or turned off. The total neutrons are then counted.

The total neutrons are a combination of the delayed neutrons from the induced fissions and prompt neutrons from spontaneous fission; multiplication amplifies both of these neutron sources. The spontaneous fission neutrons are primarily from ^{244}Cm. Hence, a key technical challenge is to produce enough interrogating neutrons so that the delayed neutron signal is similar in strength to the spontaneous fission signal.

In the safeguards context, an isotopic californium source or a neutron generator are commonly spent for active neutron interrogation. The strongest californium source commercially available has a source intensity of ~1 x 10^{10} n/s; this is at least an order of magnitude too weak. With respect to neutrons generators, a generator that uses a mixture of deuterium and tritium (DT) provides the most cost effective source of neutrons. From discussions with subject matter experts it is expected that present neutron generator technology can be spent to produce ~1 x 10^{13} n/s.[26] Hence, the neutrons source for a delayed neutron system needs to be a DT neutron generator.

However, since the neutrons from a DT generator are born with an energy of 14 MeV, there is a concern that fission in ^{238}U may hinder the ability to detect the fissile content since ^{238}U represents ~96% +/- 2% of the actinides for all commercial spent fuel assemblies. This is particularly important since ^{235}U and ^{238}U produce roughly the same number of delayed neutrons per gram when the interrogating neutron energy is above 1 MeV. In the context of quantifying Pu in spent fuel, it is important that the fission of ^{238}U not dominate the delayed neutrons signal. In other words, the instrument needs to be designed to primarily measure fissile mass (^{235}U, ^{239}Pu and ^{241}Pu).

In order to determine how strong the neutron generator needs to be and to determine how significant fissions in ^{238}U are, a MCNPX model was created. In Fig. 1, a horizontal cross section of the detector is illustrated. The fuel is depicted in the center. Moving outward from the center, surrounding the pins there may be water, borated water or air. Continuing to move further out, there is a region colored in yellow that is 10 cm across; this region

was arbitrarily dimensioned to provide room for a neutron generator. Inside of this region there are 10 annuli. These were included so that various materials could be inserted for the purpose of lowering the average energy of the neutrons incident upon the spent fuel (spectrum tailoring). The 14 MeV neutron source is in the outermost of the annuli. Exterior to this is a thin layer of cadmium. Beyond the cadmium liner is a large volume of polyethylene inside of which are embedded 36 fission chambers (93% ^{235}U).[27]

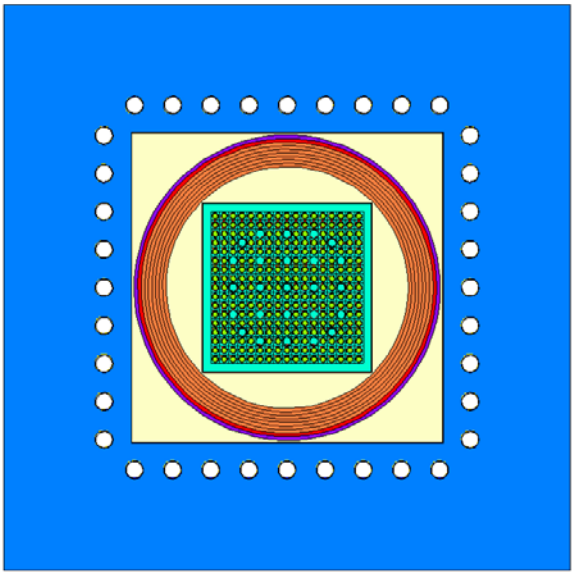

Fig. 1. Horizontal cross section of the delayed neutron detector – a description of the material in the cross section is given in the text.

To determine the performance of the detector depicted in Fig. 1, the following timing was spent: (1) neutron generator emits neutrons from 0 to 0.9 s, (2) pause of 0.1s to assure that no neutrons from the generator are counted during the delayed neutron counting interval, (3) count for 1.0 second, (4) repeat steps 1 to 3 for a total count time of 300 s. In order to determine how much the fission of ^{238}U contributes to the delayed neutron count rate, the tagging feature in MCNPX was spent. With all the annuli filled with air (no spectrum tailoring) and with a 30 GWd/tU, 3% initial enrichment and 5 years cooled assembly in the detector; the count rate during the delayed neutron counting interval was quantified. The counts from ^{235}U, ^{238}U and ^{239}Pu were all within a few percent of 30% for the case of the assembly in water. These are encouraging results since the 30% contribution of ^{238}U can be reduced with spectrum tailoring and since ^{238}U doesn't vary much among assemblies so the contribution of ^{238}U is similar to a background signal. For all the runs reported here, the outer two annuli (total of 1 cm across) were filled with tungsten while the remaining annuli (total of 3 cm across) were filled with iron. Hence, some spectrum tailoring will occur and the interrogating source will be boosted by (n, 2n) reactions. Note that no effort was made to optimize the spectrum tailoring or the boost in the interrogating source from (n, 2n) reactions.

From the modeling done to date, an estimate of the intensity needed from the neutron generator can be made. For a 45 GWd/tU, 4% initial enrichment and 5 yrs cooled assembly, the source intensity is ~4 x 10^8 neutrons/s from ^{244}Cm. Considering the following: multiplication in the assembly is nearly 2 when in water, and the detector has an efficiency of ~0.4% to 2 MeV neutrons produced anywhere in the fuel, the total background count rate is expected to be ~3 x 10^6 counts/s. In order for the delayed neutron count rate to have the same magnitude, the neutron generator would need to emit ~1 x 10^{12} neutrons/s.[27]

VI. DIFFERENTIAL DIE-AWAY

The Differential Die-away Technique (DDT) is an active interrogation technique that measures the fissile content of the fuel. The Pu content is emphasized since on a per gram basis, the combined impact from ^{239}Pu and ^{241}Pu emit ~1.6 times as many neutrons as ^{235}U. For this determination the ratio of ^{239}Pu to ^{241}Pu was taken for a ~40 GWd/tU, 4% enriched, 5 years cooled assembly.

A cross section of the detector spent in the research presented here is depicted in Fig. 2. With the DDT a measurement begins with the burst of 14 MeV neutrons from a DT neutron. The primary detectors only detect neutrons above 1 eV since they are wrapped in cadmium and borated rubber. Data is only taken after the neutrons from the neutron generator have slowed down such that virtually all the neutrons are below 1 eV . After this time the neutrons are so slow that they cannot penetrate the Cd and borated rubber, then the neutrons that are detected from the spent fuel are primarily from two sources: (1) prompt neutrons resulting from induced fissions for which the inducing neutrons were produced initially by the neutron generator, and (2) spontaneous fission neutrons from curium and any multiplication they cause. The signal that is proportional to the fissile content is the prompt neutron signal produced by fissions which are induced by the thermal neutrons from the burst. A 100-Hz repetition rate is representative for a DDT system.[28]

In Fig. 2, a horizontal cross section of the detector is illustrated. The fuel is depicted in the center. Moving outward from the center, surrounding the pins there may be water, borated water or air. Continuing to move further out, there is a region colored white that is 10 cm across; this region was arbitrarily dimensioned to provide room for a neutron generator. Next to the generator is a 1 cm thick polyethylene layer. Then a 10 cm lead region is needed in order to protect the ^{3}He tubes from the gamma emission from the fuel. Outside the four corners of the lead are the detector blocks made up of ^{3}He tubes located inside of

polyethylene slabs. The slabs are wrapped in polyethylene and borated rubber. The large yellow area is carbon and the exterior layer all around the detector is iron. No effort was made to optimize the number of tubes.

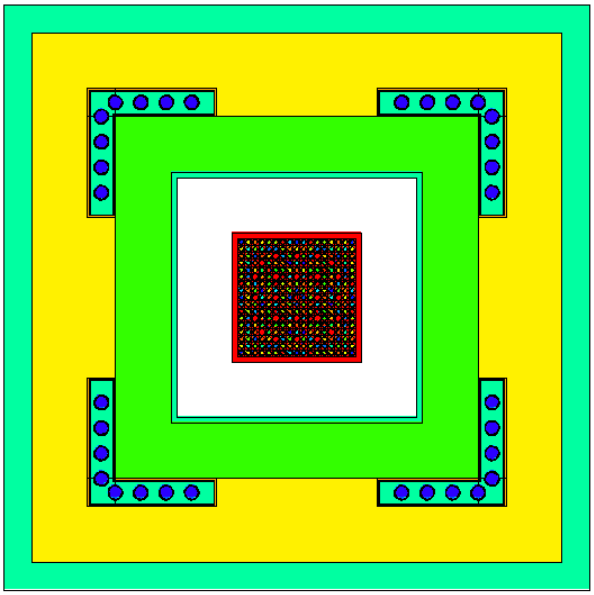

Fig. 2. Horizontal cross section of the detector – a description of the material in the cross section is given in the text.

In Fig 3, the time response of the detector is illustrated. The black data points depict the number of counts in all the ^{3}He tubes as a function of time when a fresh $^{238}UO_2$ assembly is in the detector. For this case all the uranium was ^{238}U so that this data could be spent to determine when the signal in the tubes was nearly zero in the absence of fissile material. The data in red is the temporal response when a realistic assembly is assayed. The integration time was selected to be from 1 to 10 ms.[28]

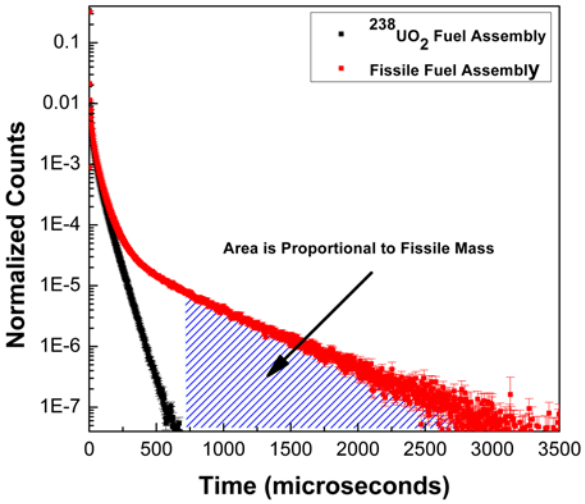

Fig. 3. The total counts in all the ^{3}He of the DDT detector are depicted as a function of time for the case when a pure $^{238}UO_2$ assembly is in the detector and when a typical spent fuel assembly was in the detector.

In Fig. 4 the count rate for 16 assemblies are given for the full range of burnup and initial enrichment in the spent fuel library. The cooling time was 5 years and the intensity of the neutron generator was 10^8 neutrons per burst or 10^{10} n/s.[28]

From the data of Fig. 4, the background in a DDT system can be compared to the DDT signal for the same assembly case spent with the delayed neutron detector. For a 45 GWd/tU, 4% initial enrichment and 5 yrs cooled assembly, the source intensity is ~4 x 10^8 neutrons/s from ^{244}Cm. Considering the following: multiplication in the assembly is nearly 2 when in water and the detector has an efficiency of ~4% to 2 MeV neutrons, the total background count rate is expected to be ~3 x 10^7 counts/s. The DDT signal for this same case is ~1 x 10^7 counts/s. Hence, a generator that produces ~3 x 10^{10} n/s is strong enough to produce a DDT signal of about the same strength as the background for this relatively high burnup case.

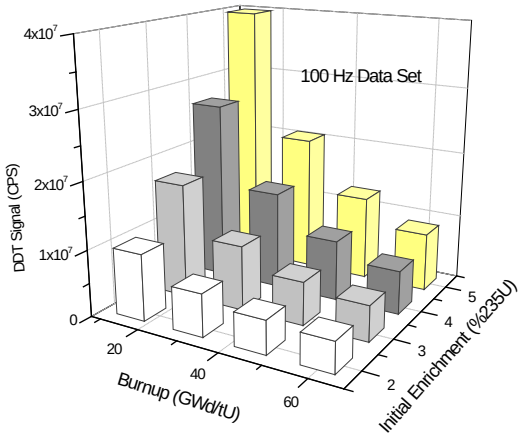

Fig 4, the count rate for 16 assemblies is given for the full range of burnup and initial enrichment in the spent fuel library for a cooling time of 5 years with a neutron generator source strength of 1 x 10^{10} n/s.

VII. NUCLEAR RESONANCE FLUORESCENCE

As stated earlier, it will be necessary to have one of the more penetrating neutron techniques in order to detect the diversion of internal pins. Furthermore, the signatures from these techniques need to measure the presence of plutonium. The six techniques best suited for this are the following: Delayed Neutrons, Differential Die-Away, Neutron Multiplicity, Passive Neutron Albedo Reactivity, ^{252}Cf Interrogation with Prompt Neutron Detection, Lead Slowing Down Spectrometer. The first 5 of these techniques measure the fissile content in the fuel which is primarily a weighted sum of ^{235}U, ^{239}Pu, and ^{241}Pu. The Lead Slowing Down Spectrometer technique independently measures the mass of ^{235}U, ^{239}Pu, and possibly ^{241}Pu.

All of the techniques listed above need more information in order to quantify elemental Pu. The techniques measuring fissile content need, at a minimum, some way to separate how much of the fissile content signal is from Pu and how much is from U. The Lead Slowing Down Spectrometer and all the techniques measuring fissile content need some means for quantifying the 3 or 4 isotopes of Pu not directly measured. Nuclear Resonance Fluorescence (NRF) is a particularly promising technique for providing exactly the information needed by the neutron techniques. It may be able to measure the absolute, or at least the relative abundances, of the key actinides of interest.[29] Since it is likely that the relative abundances can be determined more accurately than the absolute and since this is the information needed by the neutrons techniques, it is most likely that NRF will be spent to determine the relative abundances in a spent fuel assay system.

NRF is an active interrogation technique. An intense photon source, most likely bremsstrahlung, illuminates the fuel causing excitation of multiple nuclei. When a given nucleus de-excites, one or more photons are emitted. Since the de-excitation occurs between definite nuclear states, discrete gamma rays are produced that are indicative of the specific isotope from which they originated. In general ~10 distinct spectral lines are expected to exist for each actinide in the 1 to 3 MeV energy range.[29] The intensity of the radiation produced is proportional to the concentration of a given isotope in the fuel.

If NRF could determine the relative mass of ^{235}U and ^{239}Pu, it would significantly help all the techniques measuring fissile content. Any additional isotopic information, particularly ^{241}Pu, would further increase the usefulness of NRF. The following 10 isotopes are of particular interest to safeguarding spent fuel: ^{235}U, ^{238}U ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{244}Cm. The cross sectional data has only been measured for the three isotopes that are underlined. Given the anticipated time and cost needed to obtain the full suite of needed nuclear data, only a rough estimate of the capability of NRF in assaying spent fuel is possible at this time.

Recently, a collaboration among the University of California at Berkeley, Lawrence Berkeley National Laboratory and Los Alamos National Laboratory has resulted in the preliminary inclusion of NRF physics into the MCNPX code. In Fig. 5 the spectrum of the photon flux calculated at a point 60 cm from a spent fuel pin is illustrated for the case of an interrogating bremsstrahlung beam with an endpoint energy of 2.0 MeV; the resolution is that of a HPGe detector. The detector is positioned to detect backscattered photons. The pin contained fission fragments and the full suite of actinides (0.5% ^{235}U). The 6 strongest peaks depicted are from ^{238}U while several of the smaller peaks are from ^{235}U. It is important to note that these are preliminary results and the intensity of the background continuum, in particular, is a subject of ongoing research.[30]

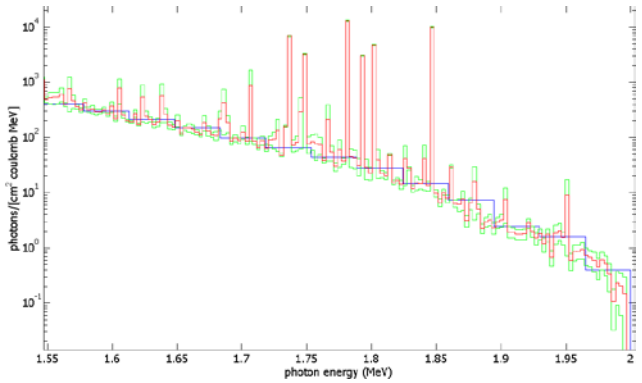

Fig. 5, Portion of the MCNPX generated spectra detected 60 cm (back scattered) from a spent fuel pin. The red line indicated the expected spectra. The difference between the red and green lines indicated the one sigma uncertainty. The blue line illustrated the spectra without NRF physics.

The blue line indicates the spectrum expected before NRF physics was added to the MCNPX code. The red line indicates the spectrum expected with NRF physics included. The difference between the green spectra and the red spectra indicates the one sigma uncertainty determined by the MCNPX code.[30]

Note that the background from the intrinsic gamma emission from the spent fuel is not included. Note that the vast majority of the photons emitted from spent fuel are emitted at much lower energies than the NRF lines. This is particularly true for the NRF lines from ^{239}Pu which are primarily in the 2 to 2.5 MeV range; the last major peak emitted by fission fragments in spent fuel is the 2.186 MeV line of ^{144}Pr.

It is also worth noting that photons in the 1 to 3 MeV energy range are much more penetrating than most of the spectral lines commonly spent in safeguards. Even though this is true, attenuation is expected to be sufficiently great so as to render the NRF signal effectively blind to pins at the center of an assembly. To quantify this point, in traversing through the diameter of 5 pins, the intensity of a 2 MeV photon is reduced by ~90%. Note that an interrogation beam is attenuated as it penetrates into the assembly to get to a nucleus. Then, after the interrogating photon excites a nucleus, the photon liberated by the de-excitation is attenuated by roughly the same amount on the way out of the assembly. The “one way” 90% estimate is clearly high since it is for a photon that goes through the diameter of each of the five pins. Most photons will go through only part of a rod and some streaming around pins will occur. Monte Carlo calculations with the diversion scenarios will quantify the sensitivity of NRF to missing pins. The main point to be made now is that it is expected that the signal from the outer pins will strongly dominate the overall signal so as to render NRF effectively blind to the inner portion of an assembly.

Since the relative masses determined by NRF are representative of the exterior pins more so than the interior pins, a slight bias is expected if no correction were made. It is anticipated that a calibration factor can be determined either through measurements or modeling. As a near-term step, modeling can indicate the magnitude of the anticipated bias.

VIII. PLUTONIUM ISOTOPIC CORRELATION

As stated earlier, none of the NDA techniques can be spent as a standalone technique to quantify the elemental mass of Pu in an assembly. Each technique measures a signature or signatures that need to be combined with other pieces of information. Some of that information will come from other instruments but some could come from a general understanding of how fuel changes in a reactor. It will be up to each regulatory body to decide what information they find to be acceptable.

In Fig. 6, the variation in the Pu mass for a full assembly is given as a function of burnup for the 5 primary isotopes of Pu for an assembly with an initial enrichment of 4% ^{235}U after a cooling time of 5 years. This data could be obtained experimentally or from burnup codes. The use of data such as that in Fig. 6 will be referred to as “Pu isotopic correlation.” The data in Fig. 6 is from assemblies in the spent fuel library.[25]

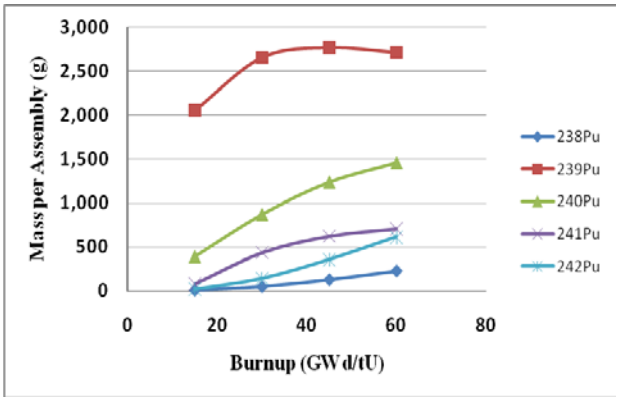

Fig. 6, the mass of the 5 main isotopes of Pu in all the assemblies in the spent fuel library are illustrated as a function of burnup for the case with an initial enrichment of 4% and a cooling time of 5 years.

If the burnup and cooling time were known, then the ratio among the 5 isotopes of Pu would be known. There are two primary means of determining the burnup with NDA. The total neutron emission from spent fuel is proportional to the burnup to the third or fourth power. Given this very strong dependence, the burnup can be accurately determined from total neutron counting. Note that if fuel were diverted, this burnup estimate would be low provided an equivalent neutron source was not added. For this reason it would be advisable to measure the burnup

by two different means. By measuring the emission from cesium and europium, the burnup on the surface of the assembly can be determined. A significant variation between the burnup determined by counting total neutrons from the entire assembly and by counting gamma emission from the outer pins would send up a flag.

Although it is possible to estimate the cooling time from the gamma and neutron data, in the context of international safeguards, it is questionable if this is necessary. Every time fuel is loaded or unloaded from the reactor, an IAEA inspector is present. Hence, the cooling time is well known. The cooling time is important in the context of Pu isotopic correlation primarily since ^{241}Pu has a 14 year half-life and ^{244}Cm has an 18 year half-life. The other isotopes of primary interest have half-lives of several thousand years or more.

IX. CONCLUSIONS

A nominally 5 year research effort has recently begun focused on quantifying elemental Pu mass in spent fuel assemblies with NDA. Nine motivations for undertaking this effort were described. Twelve NDA techniques capable of quantify different signatures emitted by spent fuel were listed. The main theme of the paper is that none of the NDA techniques listed, working in isolation, can quantify elemental Pu in assemblies. Since determining elemental Pu in assemblies is the primary goal, the research plan is to integrate a few techniques together. The first two years of the five year effort is primarily a Monte Carlo (MCNPX) based effort to quantify how well each of the twelve NDA techniques is expected to perform individually and in a system. At the end of two years a system or two will be identified. In the later three years, instruments will be fabricated and spent fuel assemblies will be measured. A plan for measuring spent fuel of the NDA systems has not been finalized and is actively seeking interested collaborators.

Preliminary results for three of the twelve NDA techniques were presented. The results for two active interrogation techniques (delayed neutron and differential die-away) indicated that the neutron generators needed to enable these techniques will not be too difficult to fabricate. The exciting potential of NRF was described; in particular how it may provide exactly the information needed by the more penetrating neutron techniques. A system that combines NRF with one of the neutron techniques could potentially determine elemental Pu mass and detect missing pins at the center of an assembly. Preliminary results of some of the first NRF spectra modeled in MCNPX were presented; this new modeling tool is in the early stages of benchmarking. The potential use of “Pu isotopic correlation” was discussed – an approach that involves quantifying the ratio among the Pu isotopes from burnup signatures.

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